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A novel approach to quantifying EPP Influences on the budgets of stratospheric NOy and ozone

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Outline

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  - CESM 2.0 Specified Dynamics (SD) WACCM
  - EPP NOy tagging approach
- Simulation description
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CESM2.0: SD-WACCM

Horizontal resolution	0.95˚x1.25˚	No. Species	228 + 33 tagged
Vertical layers	88 (0-140 km)	Kinetic rates	JPL-15
Boundary Layer	CLUBB	Sulfate SAD	Interactive with MAM
Microphysics	MG2.0	ICE SAD	MG2.0
Radiation	RRTMG	Solar Irradiance	SOLARIS-CMIP6
Aerosols	MAM-4	GHG / Halogens	Meinshausen, 2016
EPP (MEE, SPE, GCR)	SOLARIS-CMIP6	Met. Fields	MERRA-2

Tagging NOy approach

$NOy = \{ N + NO + NO_2 + NO_3 + 2N_2O_5 + HNO_3 + HO_2NO_2 + ORGNOY + NH_4NO_3 \}$

- Additional chemical and photochemical reactions are added to the chemical mechanism. These reaction **do not affect original species abundances**
- However, they use the abundance and rate constants information of the original mechanism, e.g.,
  - **$NO_2 + O_3 \rightarrow NO_3 + O_2$**
  - **$XNO_2 + O_3 \rightarrow XNO_3 + O_3$** (does not affect ozone)
- Tagged species to undergo wet and dry deposition processes
- Stratospheric heterogeneous processes included, e.g.,
  - **$XClONO_2$** + HCl on STS Aerosol
  - Denitrification of **$XHNO_3$** on NAT via settling

Symbol	Composition	Symbol	Composition
XN	N	XPAN	$CH_3CO_3NO_2$
XN2D	$N(^2D)$	XMPAN	$CH_2CCH_3CO_3NO_2$
XNO	NO	XONITR	$C_4H_7NO_4$
XNO2	NO_2	XHONITR	$C_4H_9NO_4$
XNO3	NO_3	XNOA	$CH_3COCH_2ONO_2$
XHNO3	HNO_3	XNTERPO2	$C_{10}H_{16}NO_5$
XNO2NO3	N_2O_5	XNTERPOOH	$C_{10}H_{17}NO_5$
XNO3NO2	N_2O_5	XPBZNIT	$C_7H_5O_3NO_2$
XHO2NO2	HO_2NO_2	XTERPNIT	$C_{10}H_{17}NO_4$
XHNO3	HNO_3	XNC4CH2OH	$C_5H_9NO_4$
		XNC4CHO	$C_5H_7NO_4$
XBRONO2	$BrONO_2$	XNC4CHO	$C_5H_7NO_4$
XCLONO2	$ClONO_2$	XALKNIT	$C_5H_{11}ONO_2$
		XISOPNITA	$C_5H_9NO_4$
XO	O	XISOPNITB	$C_5H_9NO_4$
XO3	O_3	XISOPNO3	$C_5H_8NO_5$
XO1D	$O(^1D)$	XISOPNOOH	$C_5H_9NO_5$
		XNC4CH2OH	$C_5H_9NO_4$

Simulation description

- 10 year simulation (1999-2008 inclusive)
- Constrained to MERRA-2 reanalysis
- HEPPA-SOLARIS CMIP6 odd-nitrogen sources are tagged at emission
  - Solar proton events (next slide)
  - Medium energy electron (30 keV and 1 MeV) precipitation
  - Galactic cosmic rays
- Auroral (~2 keV) sources not tagged (NO is produced via E-region ion chemistry)
- ‘Branch’ simulation initialized with model state in January 2004 with CMIP6 odd-nitrogen sources switched off

GCR Production… Jackman et al., JGR, 2009

Table 1. Largest Daily NO_y Production From Solar Proton Events Between 1 January 1995 and 31 December 2004, Given in Chronological Order[a]

Date of SPE	Computed NO_y Daily Production In Middle Atmosphere (gigamoles[b])
14 July 2000	1.3
15 July 2000	4.5
9 November 2000	3.6[c]
10 November 2000	0.7[c]
25 September 2001	1.4
26 September 2001	1.3
5 November 2001	2.0
6 November 2001	3.0
23 November 2001	0.8
24 November 2001	2.0
21 April 2002	0.5
22 April 2002	0.4
28 October 2003	0.8
29 October 2003	3.9
30 October 2003	0.9
3 November 2003	0.4

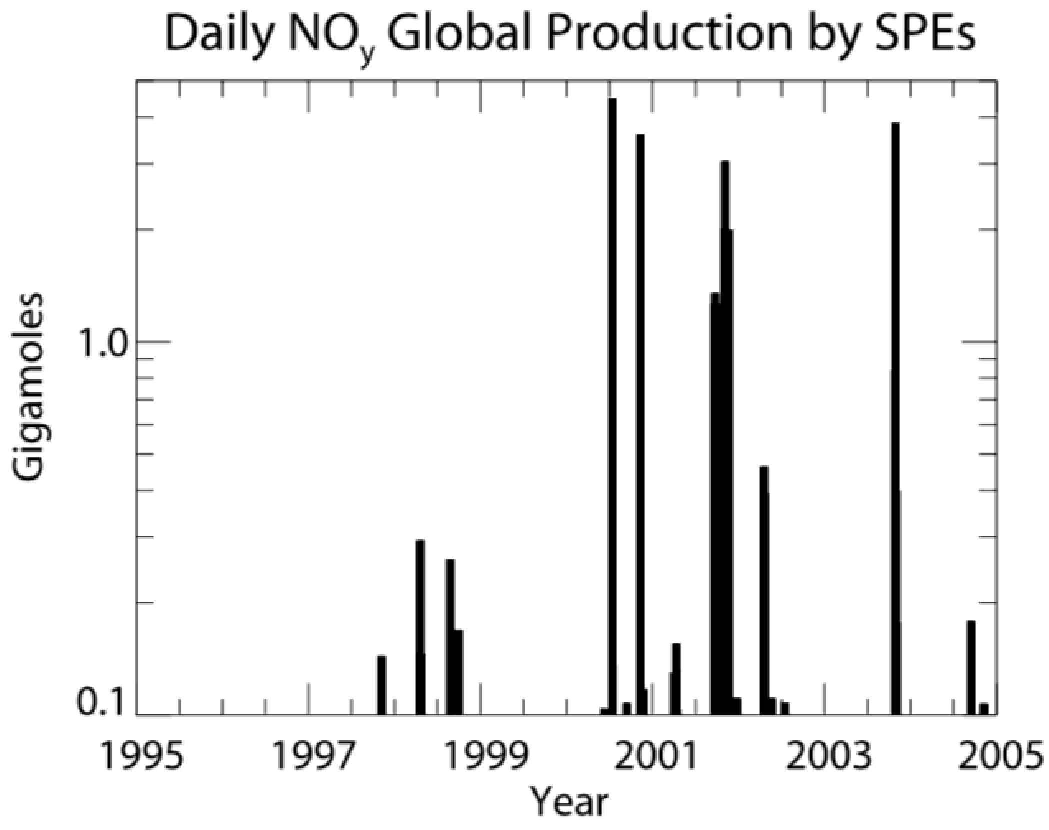
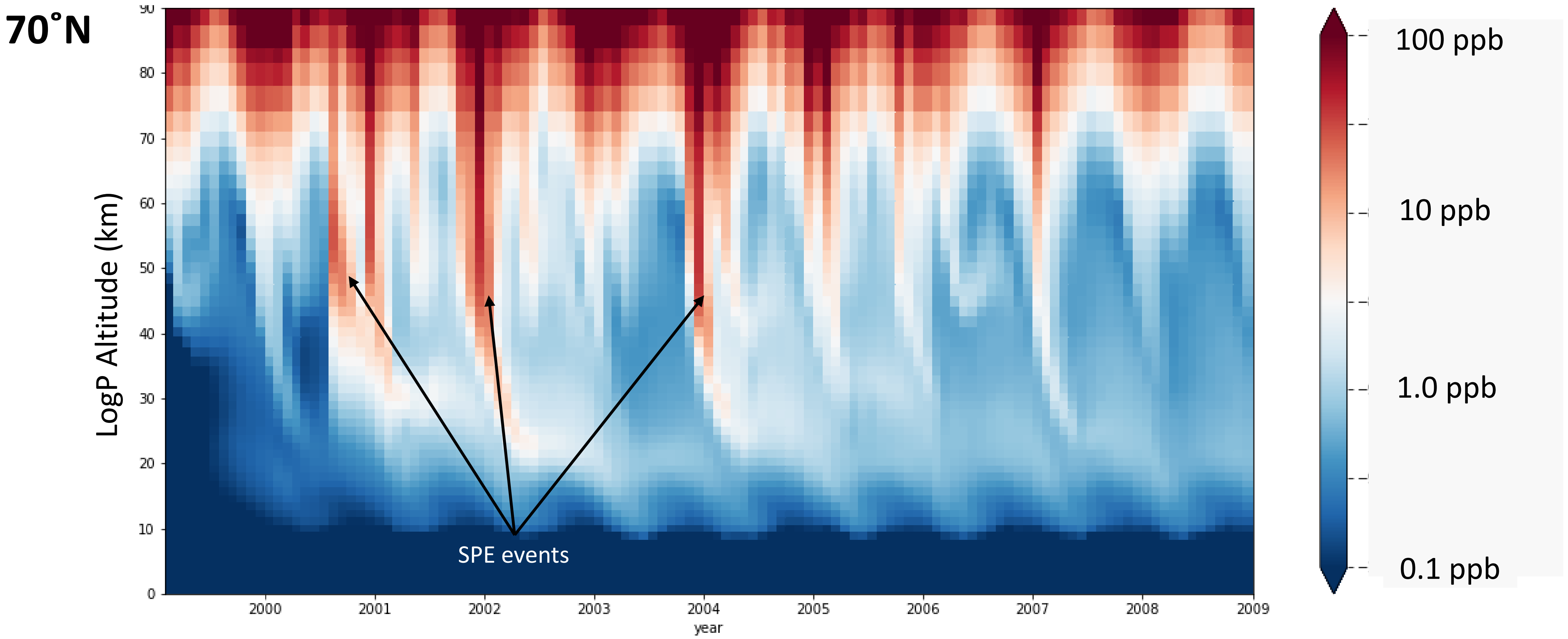


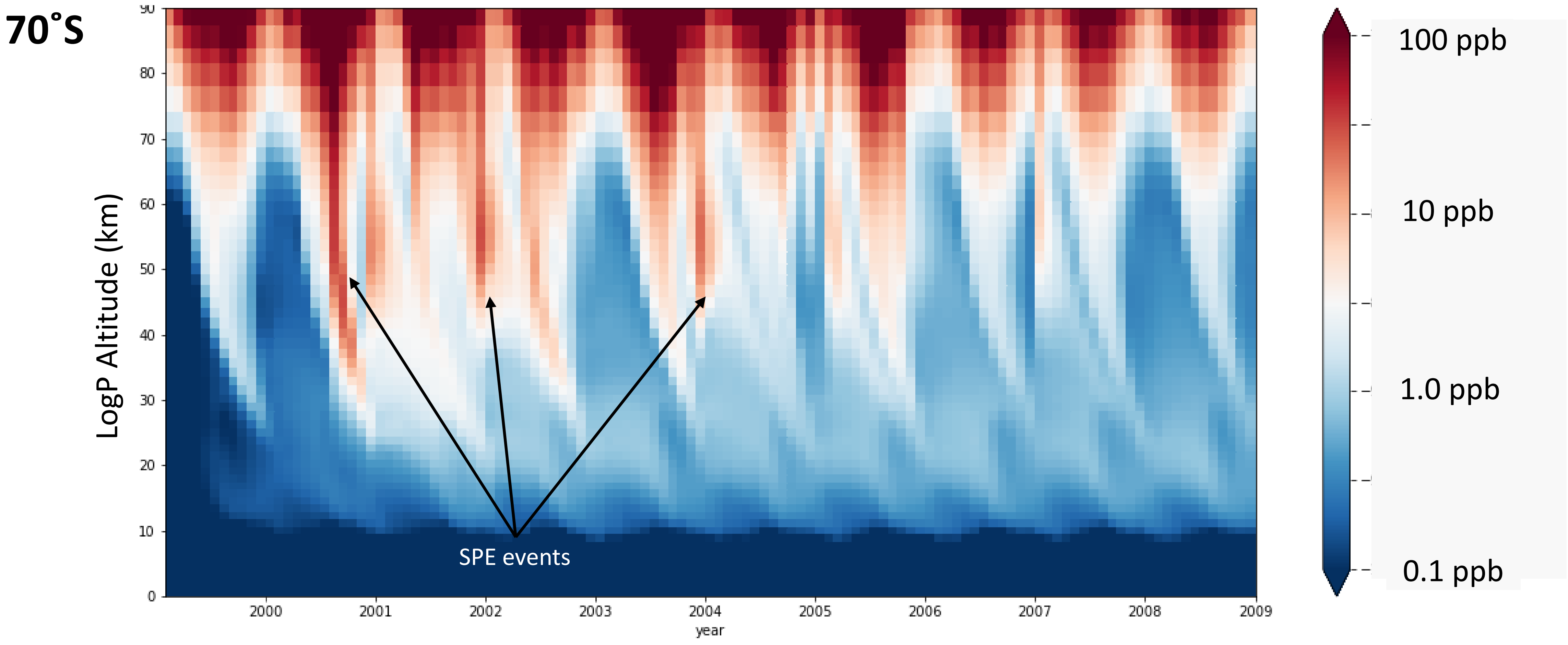
Figure 1. Daily column NO_y production in gigamoles (6.02×10^{32} molecules) as a function of time for 1 January 1995 to 31 December 2004.

EPP-NOy wintertime descent (NH)

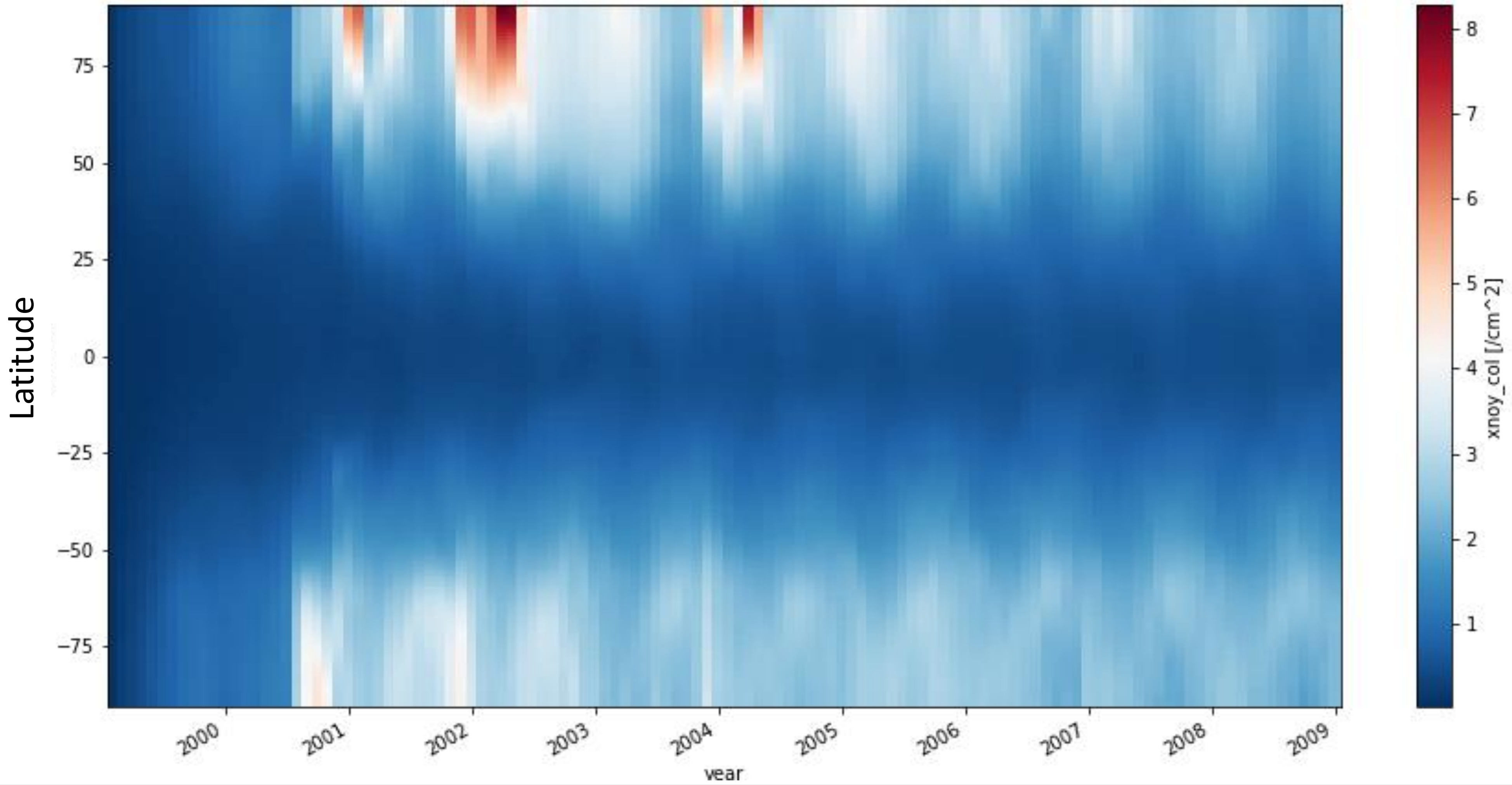


$NOy = N + NO + NO_2 + NO_3 + 2N_2O_5 + HNO_3 + HO_2NO_2 + ORGNOY + NH_4NO_3$

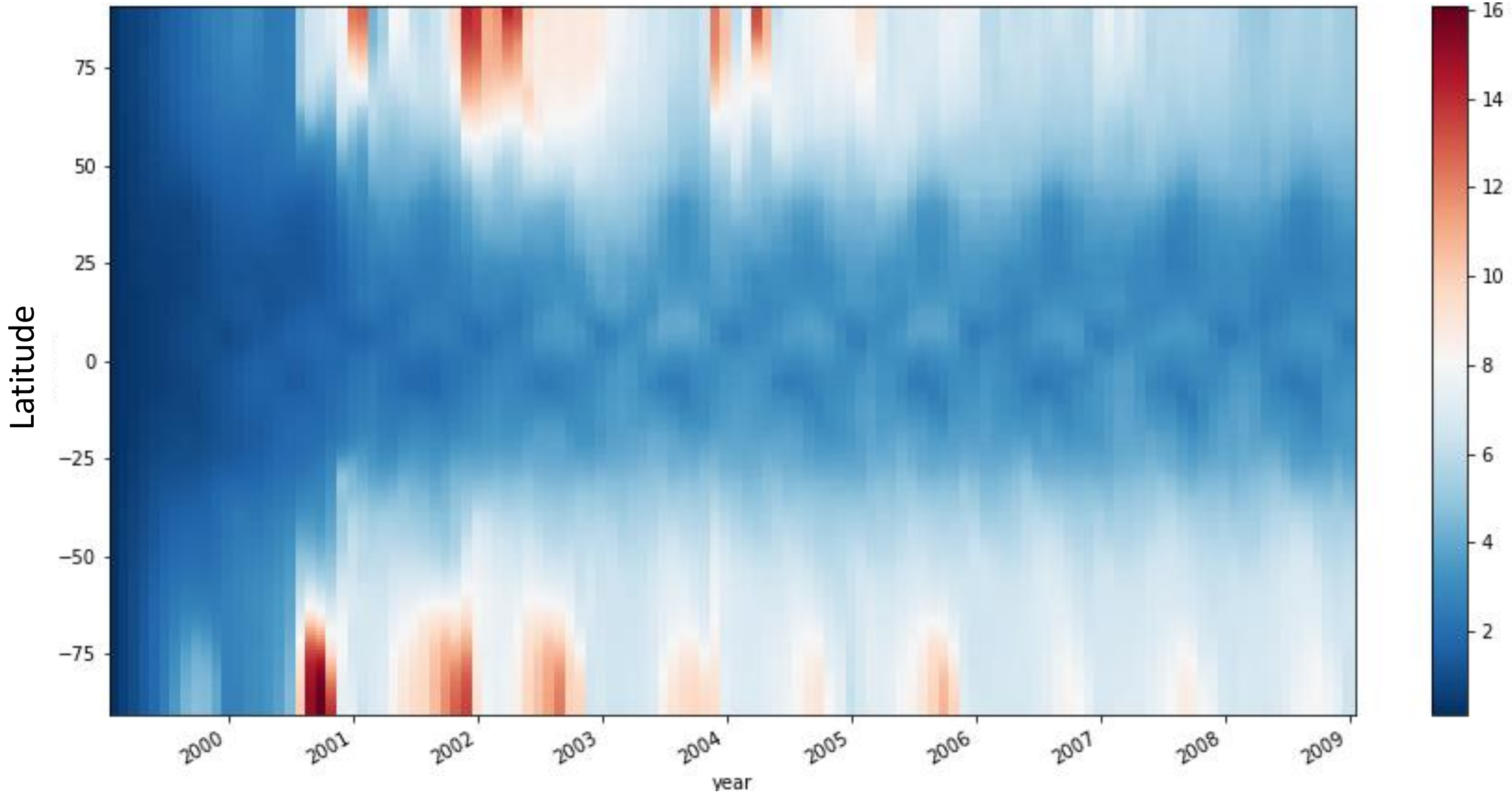
EPP-NOy wintertime descent (SH)



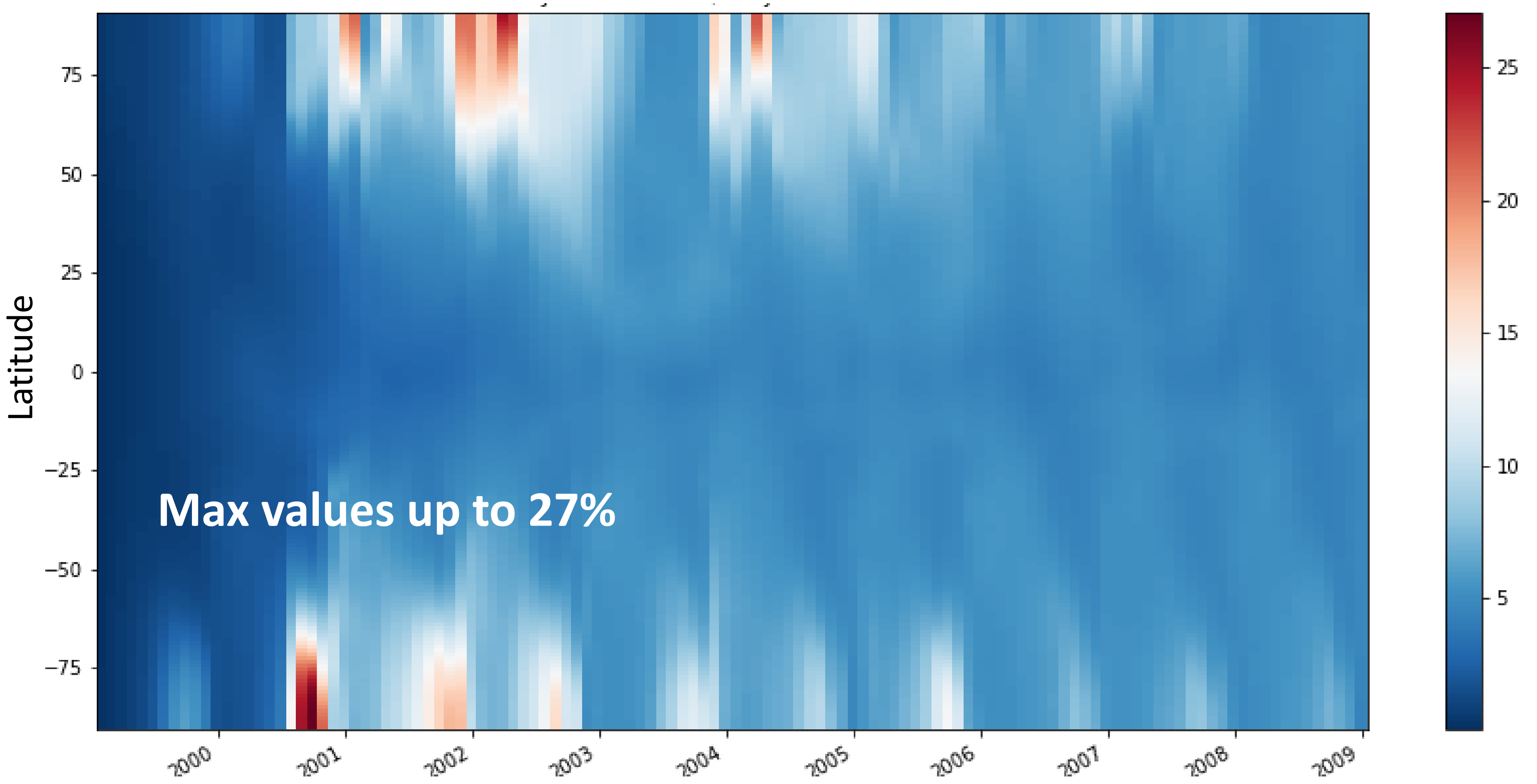
EPP-NOy total column



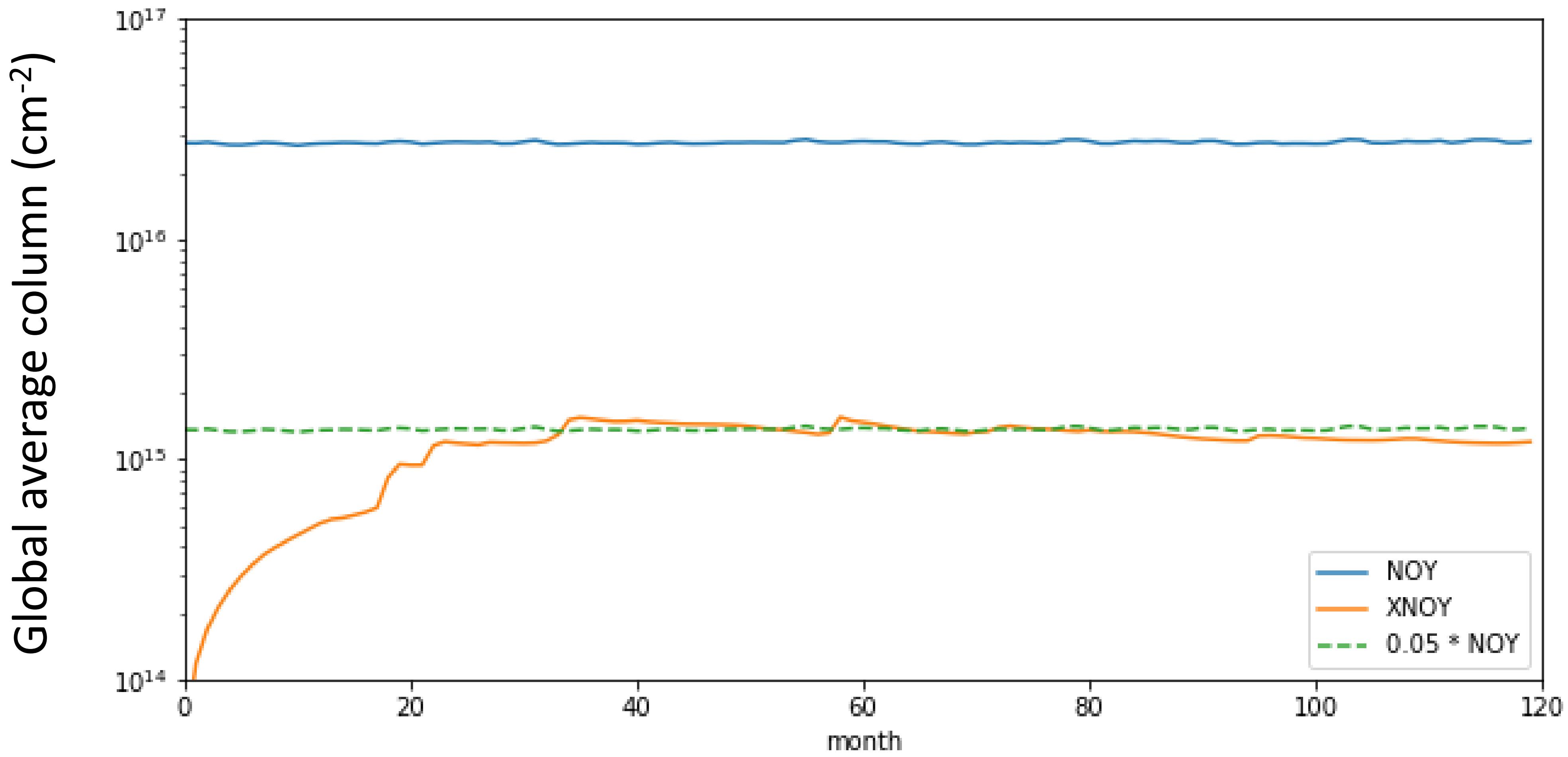
EPP-NOy to NOy total column ratio (%)



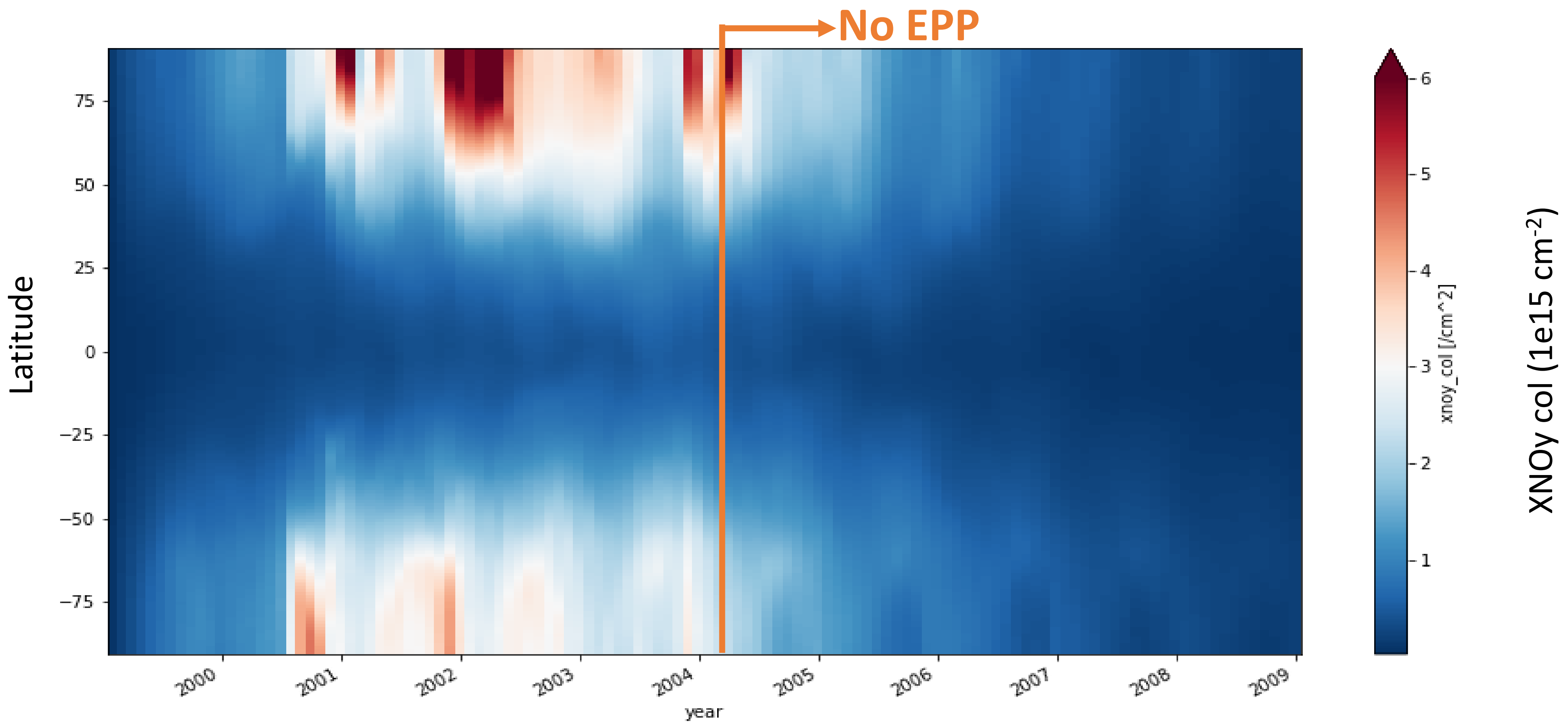
EPP- NOy to NOy partial column ratio <92 hPa (%)



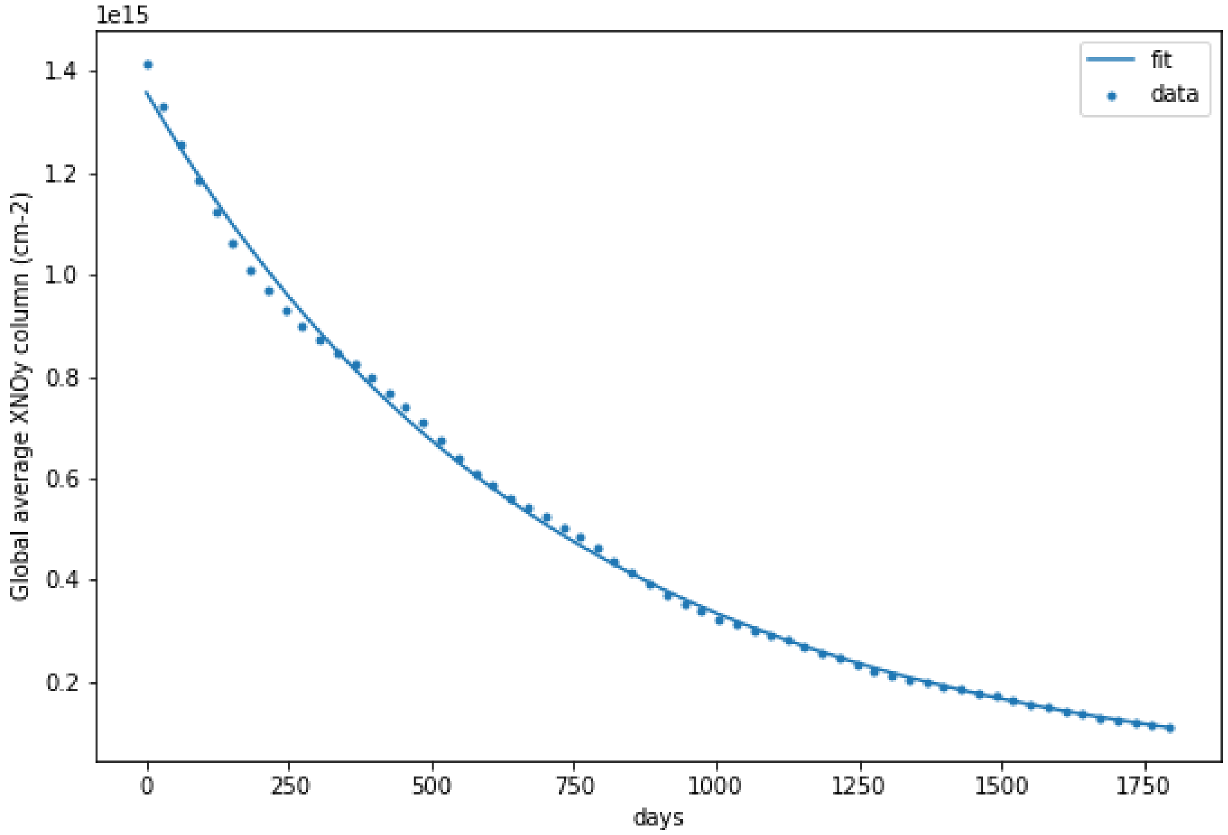
Global mean NOy column



EPP-NOy total column – no EPP after 2004



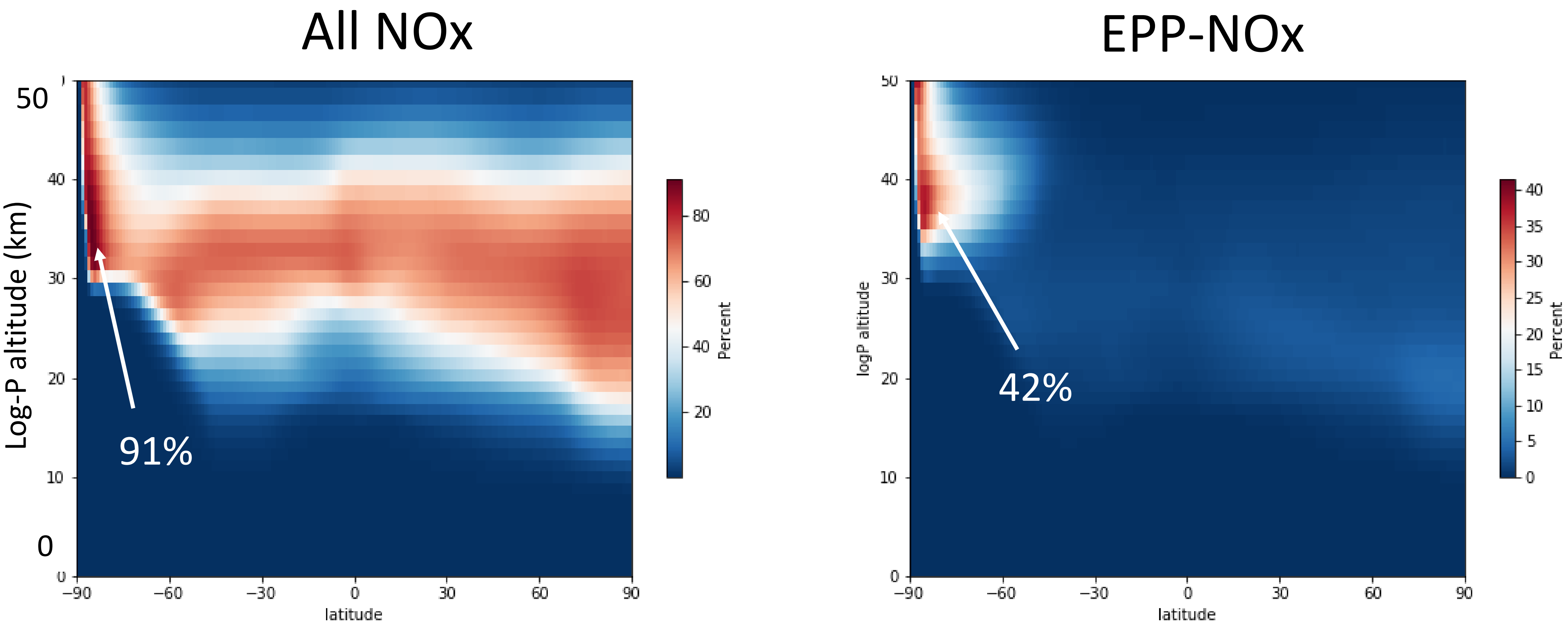
Global mean EPP-NOy lifetime



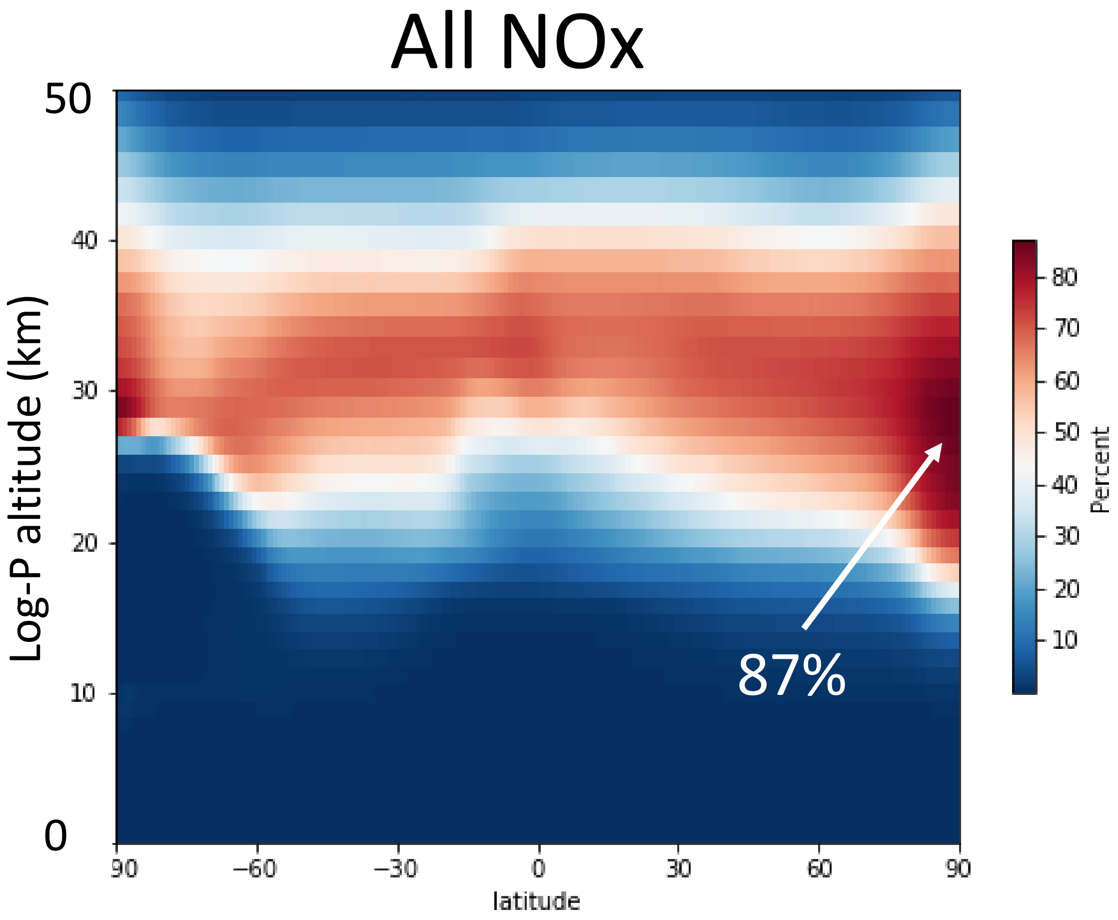
fit $N(t) = N_0 \, e^{-t/\tau}$

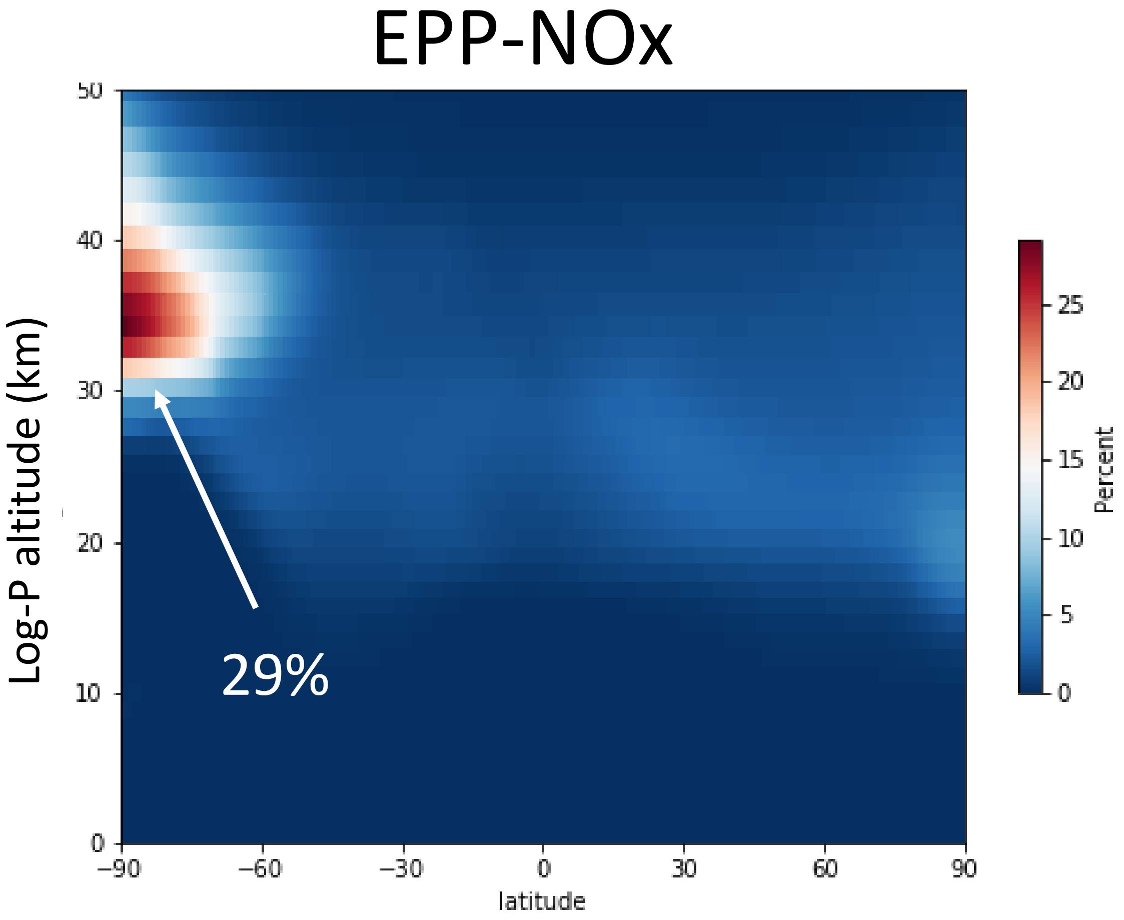
mean lifetime τ = 23.81 months

2*R(NO_2+O) / all odd-oxygen loss in Aug. 2003



2*R(NO_2+O) / all odd-oxygen loss in Sep. 2003





Summary

- New tagging scheme allows tracing of NOx by their sources and quantification of their contribution to total NOy.
- SPE+MEE+ GCR EPP contributes approximately 5% of the total globally averaged NOy column
  - Up to 20% at high latitudes
  - Up to almost 30% of the column above the tropopause
- Mean lifetime of EPP-NOy in atmosphere is ~24 months
- Contribution of EPP-NOy to total stratospheric ozone loss varies with season and height – can be up to 40%
- Values are a lower limit since it excludes auroral electron sources